Force-velocity relation and density profiles for biased diffusion in an adsorbed monolayer

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Abstract

In this paper, which completes our earlier short publication [Phys. Rev. Lett. **84**, 511 (2000)], we study dynamics of a hard-core tracer particle (TP) performing a biased random walk in an adsorbed monolayer, composed of mobile hard-core particles undergoing continuous exchanges with a vapor phase. In terms of an approximate approach, based on the decoupling of the third-order correlation functions, we obtain the density profiles of the monolayer particles around the TP and derive the force-velocity relation, determining the TP terminal velocity, V_{tr}, as the function of the magnitude of external bias and other system's parameters. Asymptotic forms of the monolayer particles density profiles at large separations from the TP, and behavior of V_{tr} in the limit of small external bias are found explicitly.

PACS numbers: 05.40-a, 66.30.Lw, 68.45.Da

1 Introduction.

When a gas or vapor is brought into contact with a clean solid surface, some portion of the gas particles becomes reversibly attached to the surface forming an adsorbed layer. Following the seminal work of Langmuir (see, e.g., Ref.[1]), equilibrium properties of such adsorbed layers have been extensively studied and a significant number of important advancements have been made. In particular, subsequent analysis included more realistic forms of intermolecular interactions or allowed for a possibility of multilayer formation. As a result, different phase transformations have been predicted and different forms of adsorption isotherms have been deduced, which are well corroborated by available experimental data (see, e.g. Refs.[1–3]).

Some work has been focused on understanding of random motion of individual molecules in adsorbed layers, which constitutes an important factor limiting their global dynamical behavior. For instance, transport processes control spreading rates of molecular films on solid surfaces [4, 5], spontaneous or forced dewetting of monolayers [6–9] or island formation [10]. Here, some approximate analytical results have been obtained for both dynamics of an isolated adatom on a corrugated surface and collective diffusion, describing spreading of macroscopic density fluctuations in interacting adsorbates being in contact with the vapor phase [11–14]. Analysis of tracer diffusion in adsorbed layers, which provides a useful information about intrinsic friction of the adsorbates, pertains mostly to two-dimensional models with forbidden particles exchanges with the vapor (see, e.g. Refs.[15–20]. Tracer diffusion in adsorbed monolayers undergoing continuous adsorption and desorption has been essentially less studied. A few available detailed studies are either devoted to an analysis of a somewhat artificial one-dimensional situation [21], or, for two-dimensional adsorbates in contact with the vapor, focus on the limit when the tracer particle perform a totally directed random walk [22]. The results for the general situation with arbitrary bias has been only briefly presented in Ref.[23].

In this paper, which completes our earlier short publication [23], we present a detailed analysis of the properties of a biased tracer diffusion in two-dimensional adsorbed monolayers undergoing continuous exchanges with the vapor. The system we consider consists of (a) a solid substrate, which is modeled in a usual fashion as a regular square lattice of adsorbtion sites; (b) a monolayer of adsorbed, mobile hard-core particles in contact with a vapor and (c) a single hard-core tracer particle (TP). The monolayer particles move randomly along the lattice by performing symmetric hopping motion between the neighboring lattice sites, which process is constrained by mutual hard-core interactions, and may desorb from and adsorb onto the lattice from the vapor with some prescribed rates dependent on the vapor pressure, temperature and the interactions with the solid substrate. In contrast, the tracer particle is constrained to move on the two-dimensional lattice only, (i.e. it can not desorb to the vapor), and is subject to a constant external force E. Hence, the TP performs a biased random walk, constrained by the hard-core interactions with the monolayer particles, and always remains

within the monolayer, probing its frictional properties.

In terms of an approximate approach of Ref.[24], based on the decoupling of the third-order tracer-particle-particle correlation functions into the product of corresponding pairwise correlations, we determine the density profiles of the monolayer particles, as seen from the stationary moving tracer, and calculate analytically the terminal velocity V_{tr} attained by the tracer particle in the limit $t \to \infty$. We show that the monolayer particles distribution is strongly inhomogeneous: the local density of the monolayer particles in front of the tracer is higher than the average, which means that the monolayer particles tend to accumulate in front of the driven tracer, creating a sort of a "traffic jam", which impedes its motion. The condensed, "traffic jam"-like region vanishes as an exponential function of the distance from the tracer. The characteristic length and the amplitude of the density relaxation function are calculated explicitly. On the other hand, past the tracer the local density is lower than the average. Here, we find that depending on whether the number of particles in the monolayer is explicitly conserved or not, the local density past the tracer may tend to the average value at large separations from the tracer in a completely different fashion: In the non-conserved case the decay of the density is described by an exponential function, while for the conserved particles number case it shows an *algebraic* dependence on the distance, revealing in the latter case especially strong memory effects and strong correlations between the particle distribution in the monolayer and the tracer position. Further on, we find that the terminal velocity of the tracer particle depends explicitly on both the excess density in the "jammed" region in front of the tracer, as well as on the density in the depleted region past the tracer. We realize that both densities are themselves dependent on the magnitude of the tracer velocity, applied external force, as well as on the rate of the adsorption/desorption processes and on the rate at which the particles can diffuse away of the tracer, which results in effective non-linear coupling between V_{tr} and E. In consequence, in the general case (for arbitrary adsorption/desorption rates and arbitrary external force), V_{tr} can be found only implicitly, as the solution of a transcendental equation relating V_{tr} to the system parameters. This equation simplifies considerably in the limit of a vanishingly small external bias; in this case we obtain a linear force-velocity relation, akin to the so-called Stokes formula, which signifies that in this limit the frictional force exerted on the tracer particle by the host medium (the adsorbed monolayer) is viscous. The corresponding friction coefficient is also determined explicitly.

We finally remark, that a qualitatively similar physical effect has been predicted recently for a different model system involving a charged particle moving at a constant speed a small distance above the surface of an incompressible, infinitely deep liquid. It has been shown in Refs.[25, 26], that the interactions between the moving particle and the fluid molecules induce an effective frictional force exerted on the particle, producing a local distortion of the liquid interface, - a bump, which travels together with the particle and increases effectively its mass. The mass of the bump, which is analogous to the jammed region appearing in our model, depends itself on the particle's velocity resulting in a non-linear coupling between the

medium-induced frictional force exerted on the particle and its velocity [25, 26].

The paper is structured as follows: In Section 2 we formulate the model and introduce basic notations. In Section 3 we write down the dynamical equations which govern the time evolution of the monolayer particles and of the tracer, and outline the decoupling procedure. Section 4 is devoted to the analytical solution of the decoupled discrete-space evolution equations in the limit $t \to \infty$ in terms of the generating function approach. In Section 5 we analyse the asymptotical behavior of the density profiles at large separations in front of and past the tracer particle. Next, in Section 6, we derive formal expressions describing the shape of the density profiles in the adsorbed monolayer and a force-velocity relation in the general case. Section 7 is devoted to the analysis of the general force-velocity relation in the limit of a vanishingly small external bias. Here, we derive an analog of the Stokes formula for biased diffusion in adsorbed monolayers undergoing continuous exchanges with the vapor phase, determine the friction coefficient and estimate, through the Einstein relation, the tracer diffusion coefficient. Finally, we conclude in Section 8 with a brief summary and discussion of our results.

2 The model

Consider a two-dimensional square lattice of adsorption sites of spacing σ, which is brought in contact with a reservoir containing identic, electrically neutral particles (vapor phase) (Fig.1), maintained at a constant pressure. We suppose that (a) the particles may leave the reservoir and adsorb onto any vacant lattice site at a fixed rate f/τ^*; (b) the adsorbed particles may move randomly along the lattice by hopping at a rate $1/4\tau^*$ to any of four neighboring sites, which process is constrained by hard-core exclusion preventing multiple occupancy, and (c) the adsorbed particles may desorb from the lattice back to the reservoir at rate g/τ^*. Both f and g are site and environment independent.

To describe the time-dependent occupancy of lattice sites, we introduce the variable $\eta(\mathbf{R})$, which may assume two values:

$$\eta(\mathbf{R}) = \begin{cases} 1, & \text{if the site } \mathbf{R} \text{ is occupied by an adsorbed particle,} \\ 0, & \text{if the site } \mathbf{R} \text{ is empty.} \end{cases}$$

Evidently, local $\eta(\mathbf{R})$ can change its value due to adsorption, desorption and constrained random hopping events, and the total number of particles in the adsorbed monolayer is not explicitly conserved due to adsorption/desorption processes. On the other hand, the mean density ρ_s of the adsorbate, $\rho_s =< \eta(\mathbf{R}) >$, approaches as $t \to \infty$ a constant value

$$\rho_s = \frac{f}{f+g}, \tag{1}$$

which relation represents the customary Langmuir adsorption isotherm [1]. We note also that in the analysis of the stationary-state behavior, we can always turn to the conserved particles number limit by setting f and g equal to zero and keeping their ratio fixed, i.e. supposing that $f/g = \rho_s/(1-\rho_s)$. This limit will correspond to the model of biased tracer diffusion in a two-dimensional hard-core lattice gas with fixed particles density ρ_s, and will allow us to check our analytical predictions against some already known results [15–20].

Further on, at $t = 0$ we introduce at the lattice origin an extra hard-core particle, whose motion we would like to follow here and whose position at time t we denote as $\mathbf{R}_{tr}$. This tracer particle - the TP, can be thought of as an external probe designed to measure the resistance offered by the monolayer particles to the external perturbance, or, in other words, to measure the intrinsic frictional properties of the adsorbate.

Now, we stipulate that the TP is different from the adsorbed particles in two aspects: first, it can not desorb from the lattice and second, it experiences an action of some external driving force, which favors its jumps into a preferential direction. Physically, such a situation may be realized, for instance, if this only particle is charged and the system is subject to a uniform electric field $\mathbf{E}$. We suppose here, for simplicity of exposition, that the external force $\mathbf{E}$ is oriented in the positive X-direction, i.e. $\mathbf{E} = (E, 0)$.

More precisely, we define the TP dynamics as follows: We suppose that the TP, which occupies the site $\mathbf{R}_{tr}$ at time t, waits an exponentially distributed time with mean[1] τ, and then attempts to hop onto one of four neighboring sites, $\mathbf{R}_{tr} + \mathbf{e}_{\boldsymbol{\nu}}$, where $\mathbf{e}_{\boldsymbol{\nu}}$ are four unit vectors of the square lattice. In what follows we adopt the notation $\nu = \{\pm 1, \pm 2\}$, where $\pm\mathbf{e_1}$ (respectively, $\pm\mathbf{e_2}$) will denote $\pm X$ (respectively, $\pm Y$) directions. Next, the jump direction is chosen according to the probablity p_ν, which is defined in a usual fashion as

$$p_\nu = \frac{\exp\left[\frac{\beta}{2}(\mathbf{E}\cdot\mathbf{e}_{\boldsymbol{\nu}})\right]}{\sum_\mu \exp\left[\frac{\beta}{2}(\mathbf{E}\cdot\mathbf{e}_{\boldsymbol{\mu}})\right]}, \tag{2}$$

where β is the reciprocal temperature, $(\mathbf{E}\cdot\mathbf{e})$ stands for the scalar product, the charge of the TP is set equal to unity and the sum with the subscript μ denotes summation over all possible orientations of the vector $\boldsymbol{e}_\mu$, that is $\mu = \{\pm 1, \pm 2\}$.

After the jump direction is chosen, the TP attempts to hop onto the target site. The hop is instantaneously fulfilled if the target site is vacant at this moment of time; otherwise, i.e., if the target site is occupied by any adsorbed particle, the jump is rejected and the TP remains at its position.

[1]We suppose that in the general case this mean time τ is different from the corresponding time τ^* associated with the monolayer particles dynamics. As a matter of fact, this should be the case merely because the TP-substrate interactions may be different from the particle-substrate ones. Varying τ we can mimic different possible situations; in particular, $\tau = 0$ corresponds to the case when the tracer simply slides along the substrate regardless of the surface corrugation.

3 Evolution equations

Let $\eta \equiv \{\eta(\mathbf{R})\}$ denote the entire set of the occupation variables, which defines the instantaneous configuration of the adsorbed particles at the lattice at time moment t. Next, let $P(\mathbf{R_{tr}}, \eta; t)$ denote the joint probability of finding at time t the TP at the site $\mathbf{R_{tr}}$ and all adsorbed particles in the configuration η. Then, denoting as $\eta^{\mathbf{r},\nu}$ a configuration obtained from η by the Kawasaki-type exchange of the occupation variables of two neighboring sites $\mathbf{r}$ and $\mathbf{r} + \mathbf{e}_{\boldsymbol{\nu}}$, and as $\hat{\eta}^{\mathbf{r}}$ - a configuration obtained from the original η by the replacement $\eta(\mathbf{r}) \to 1 - \eta(\mathbf{r})$, which corresponds to the Glauber-type flip of the occupation variable due to the adsorption/desorption events, we have that the time evolution of the configuration probability $P(\mathbf{R_{tr}}, \eta; t)$ obeys the following master equation:

$$\begin{aligned}
\partial_t P(\mathbf{R_{tr}}, \eta; t) &= \frac{1}{4\tau^*} \sum_{\mu} \sum_{\mathbf{r} \neq \mathbf{R_{tr}} - \mathbf{e}_{\boldsymbol{\mu}}, \mathbf{R_{tr}}} \Big\{ P(\mathbf{R_{tr}}, \eta^{\mathbf{r},\mu}; t) - P(\mathbf{R_{tr}}, \eta; t) \Big\} \\
&+ \frac{1}{\tau} \sum_{\mu} p_{\mu} \Big\{ (1 - \eta(\mathbf{R_{tr}}))\, P(\mathbf{R_{tr}} - \mathbf{e}_{\boldsymbol{\mu}}, \eta; t) - (1 - \eta(\mathbf{R_{tr}} + \mathbf{e}_{\boldsymbol{\mu}}))\, P(\mathbf{R_{tr}}, \eta; t) \Big\} \\
&+ \frac{g}{\tau^*} \sum_{\mathbf{r} \neq \mathbf{R_{tr}}} \Big\{ (1 - \eta(\mathbf{r}))\, P(\mathbf{R_{tr}}, \hat{\eta}^{\mathbf{r}}; t) - \eta(\mathbf{r}) P(\mathbf{R_{tr}}, \eta; t) \Big\} \\
&+ \frac{f}{\tau^*} \sum_{\mathbf{r} \neq \mathbf{R_{tr}}} \Big\{ \eta(\mathbf{r}) P(\mathbf{R_{tr}}, \hat{\eta}^{\mathbf{r}}; t) - (1 - \eta(\mathbf{r}))\, P(\mathbf{R_{tr}}, \eta; t) \Big\},
\end{aligned} \tag{3}$$

where the subscript $\mathbf{r} \neq \mathbf{R_{tr}}$ under the summation symbol signifies that summation extends over all lattice sites except for the site occupied at this time moment by the TP.

Now, the instantaneous velocity $V_{tr}(t)$ of the TP can be obtained by multiplying both sides of Eq.(3) by $(\mathbf{R_{tr}} \cdot \mathbf{e_1})$ and summing over all possible configurations $(\mathbf{R_{tr}}, \eta)$. This results in the following exact equation determining the TP velocity:

$$V_{tr}(t) \equiv \frac{d}{dt} \sum_{\mathbf{R_{tr}}, \eta} (\mathbf{R_{tr}} \cdot \mathbf{e_1}) = \frac{\sigma}{\tau} \Big\{ p_1 \Big(1 - k(\mathbf{e_1}; t) \Big) - p_{-1} \Big(1 - k(\mathbf{e_{-1}}; t) \Big) \Big\}, \tag{4}$$

where

$$k(\boldsymbol{\lambda}; t) \equiv \sum_{\mathbf{R_{tr}}, \eta} \eta(\mathbf{R_{tr}} + \boldsymbol{\lambda}) P(\mathbf{R_{tr}}, \eta; t) \tag{5}$$

is the probability of having at time t an adsorbed particle at position $\boldsymbol{\lambda}$, defined in the frame of reference moving with the TP. Evidently, $k(\boldsymbol{\lambda})$ can also be interpreted as the density profile in the adsorbed monolayer as seen from the moving TP.

Equation (4) signifies that the instantaneous velocity of the TP is dependent on the monolayer particles density in the immediate vicinity of the tracer. If the monolayer is perfectly stirred, or, in other words, if $k(\boldsymbol{\lambda}) = \rho_s$ everywhere, (which implies immediate

decoupling of $\mathbf{R_{tr}}$ and η), one would obtain from Eq.(4) a trivial mean-field result

$$V_{tr}^{(0)} = (p_1 - p_{-1})(1-\rho_s)\frac{\sigma}{\tau}, \tag{6}$$

which states that the only effect of the medium on the TP dynamics is that its jump time τ gets merely renormalized by a factor $(1-\rho_s)^{-1}$, which represents the inverse concentration of voids in the monolayer; note that $(1-\rho_s)/\tau$ defines simply the mean frequency of successful jump events. However, the situation appears to be more complicated and, as we proceed to show, $k(\boldsymbol{\lambda})$ is different from the equilibrium value ρ_s everywhere, except for $|\boldsymbol{\lambda}| \to \infty$. This means that the TP strongly perturbs the particles distribution in the monolayer - it is no longer uniform and some non-trivial stationary density profiles emerge. Moreover, $k(\boldsymbol{\lambda})$ appears to be dependent on the TP velocity which results ultimately in a non-linear coupling between $V_{tr}(t)$ and density profiles.

Now, in order to calculate the instantaneous mean velocity of the TP we have to determine the mean particles density at the neighboring to the TP sites $\mathbf{R_{tr}} + \mathbf{e_{\pm 1}}$, which requires, in turn, computation of the density profile $k(\boldsymbol{\lambda})$ for arbitrary $\boldsymbol{\lambda}$. The latter can be found from the master equation (3) by multiplying both sides by $\eta(\mathbf{R_{tr}})$ and performing the summation over all configurations $(\mathbf{R_{tr}}, \eta)$. In doing so, we find the following set of equations (see Appendix A for more details):

$$\begin{aligned} 4\tau^* \partial_t k(\boldsymbol{\lambda};t) &= \sum_{\mu} (\nabla_\mu - \delta_{\boldsymbol{\lambda},\mathbf{e}_{\boldsymbol{\mu}}} \nabla_{-\mu}) k(\boldsymbol{\lambda};t) - 4(f+g)k(\boldsymbol{\lambda};t) + 4f \\ &+ \frac{4\tau^*}{\tau} \sum_{\mathbf{R_{tr}},\eta} \sum_{\mu} p_\mu \left(1 - \eta(\mathbf{R_{tr}} + \mathbf{e}_{\boldsymbol{\mu}})\right) \nabla_\mu \eta(\mathbf{R_{tr}} + \boldsymbol{\lambda}) P(\mathbf{R_{tr}}, \eta; t), \end{aligned} \tag{7}$$

where ∇_μ denotes the ascending finite difference operator of the form

$$\nabla_\mu f(\boldsymbol{\lambda}) \equiv f(\boldsymbol{\lambda} + \mathbf{e}_\mu) - f(\boldsymbol{\lambda}), \tag{8}$$

and

$$\delta_{\mathbf{r},\mathbf{r}'} = \begin{cases} 1, & \text{if the site } \mathbf{r} = \mathbf{r}', \\ 0, & \text{otherwise.} \end{cases}$$

The Kroneker-delta term $\delta_{\boldsymbol{\lambda},\mathbf{e}_{\boldsymbol{\mu}}}$ signifies that the evolution of the pair correlations, Eq.(7), proceeds differently at large separations and at the immediate vicinity of the TP, because of its asymmetric hopping rules, Eq.(2) (see for more details the points (a) and (b) in the Appendix A).

Note next that the contribution in the second line in Eq.(7), associated with the TP biased diffusion, is non-linear with respect to the occupation numbers such that the pair correlation function gets effectively coupled to the evolution of the third-order correlations of the form

$$T(\boldsymbol{\lambda}, \mathbf{e}_{\boldsymbol{\mu}}; t) \equiv \sum_{\mathbf{R_{tr}},\eta} \eta(\mathbf{R_{tr}} + \boldsymbol{\lambda}) \eta(\mathbf{R_{tr}} + \mathbf{e}_{\boldsymbol{\mu}}) P(\mathbf{R_{tr}}, \eta; t). \tag{9}$$

That is, Eq.(7) is not closed with respect to the pair correlations but rather represents a first equation in the infinite hierachy of coupled equations for higher-order correlation functions. One faces, therefore, the problem of solving an infinite hierarchy of coupled differential equations and needs to resort to an approximate closure scheme.

We proceed along the lines suggested in Ref.[23] and apply the simplest non-trivial closure approximation, based on the decoupling of the third-order correlation functions into the product of pair correlations. More precisely, we assume that for $\boldsymbol{\lambda} \neq \mathbf{e}_{\boldsymbol{\mu}}$ the third-order correlation can be written down in the following form

$$\begin{aligned}
& \sum_{\mathbf{R_{tr}},\eta} \eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})\eta(\mathbf{R_{tr}}+\mathbf{e}_{\boldsymbol{\mu}})P(\mathbf{R_{tr}},\eta;t) \\
\approx & \left(\sum_{\mathbf{R_{tr}},\eta} \eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})P(\mathbf{R_{tr}},\eta;t)\right)\left(\sum_{\mathbf{R_{tr}},\eta} \eta(\mathbf{R_{tr}}+\mathbf{e}_{\boldsymbol{\mu}})P(\mathbf{R_{tr}},\eta;t)\right) \\
= & \; k(\boldsymbol{\lambda};t)k(\mathbf{e}_{\boldsymbol{\mu}};t), \qquad (10)
\end{aligned}$$

i.e. the joint probability of having at time moment t one adsorbed particle in the immediate vicinity of the TP, at position $\mathbf{e}_{\boldsymbol{\mu}}$, and another particle at position $\boldsymbol{\lambda}$, is represented as the product of the corresponding pairwise probabilities. We hasten to remark that the approximate closure of the evolution equations in Eq.(10) has been already employed for studying related models of biased tracer diffusion in hard-core lattices gases and has been shown to provide quite an accurate description of both the dynamical and stationary-state behavior. The decoupling in Eq.(10) has been first introduced in Ref.[24] to determine the properties of a driven tracer diffusion in a one-dimensional hard-core lattice gas with a conserved number of particles, i.e. without an exchange of particles with the reservoir. Extensive numerical simulations performed in Ref.[24] have demonstrated that such a decoupling provides quite a plausible approximation for the model under study. Moreover, rigorous probabilistic analysis of Ref.[27] has shown that for this model the results based on the decoupling scheme in Eq.(10) are exact. Furthermore, the same closure procedure has been recently applied to describe spreading kinetics of a hard-core lattice gas from a reservoir attached to one of the lattice sites [5]. Again, a very good agreement between the analytical predictions and numerical results has been found. Next, the decoupling in Eq.(10) has been used in a recent analysis of a biased tracer dynamics in a one-dimensional model of adsorbed monolayer in contact with a vapor phase [21], i.e. a one-dimensional version of the model to be studied here. Also in this case an excellent agreement has been observed between the analytical predictions and Monte Carlo simulations data [21]. Besides, as we have already mentioned in Ref.[23], the closure of the hierarchy of the evolution equations in Eq.(10) allows us to reproduce in the limit $f, g = 0$ and $f/g = const$ (conserved particles number limit) the results of Refs.[16] and [17], which are known (see e.g. Ref.[15]) to provide a very good approximation for the tracer diffusion coefficient in two-dimensional hard-core lattice gases with arbitrary particle density.

Using the approximation in Eq.(10), we can rewrite Eq.(7) in the following closed form

$$4\tau^* \partial_t k(\boldsymbol{\lambda}; t) = \tilde{L} k(\boldsymbol{\lambda}; t) + 4f, \tag{11}$$

which holds for all $\boldsymbol{\lambda}$, except for $\boldsymbol{\lambda} = \{\mathbf{0}, \pm\mathbf{e_1}, \pm\mathbf{e_2}\}$. One the other hand, for these special sites $\boldsymbol{\lambda} = \mathbf{e}_\nu$ with $\nu = \{\pm 1, \pm 2\}$ we find

$$4\tau^* \partial_t k(\mathbf{e}_{\boldsymbol{\nu}}; t) = (\tilde{L} + A_\nu) k(\mathbf{e}_{\boldsymbol{\nu}}; t) + 4f, \tag{12}$$

where $\tilde{L}$ is the operator

$$\tilde{L} \equiv \sum_\mu A_\mu \nabla_\mu - 4(f+g), \tag{13}$$

and the coefficients A_μ are defined by

$$A_\mu(t) \equiv 1 + \frac{4\tau^*}{\tau} p_\mu (1 - k(\mathbf{e}_{\boldsymbol{\mu}}; t)). \tag{14}$$

Now, several comments about Eqs.(11) and (12) are in order. First of all, let us note that Eq.(12) represents, from the mathematical point of view, the boundary conditions for the general evolution equation (11), imposed on the sites in the immediate vicinity of the TP. As we have noticed already, Eqs.(11) and (12) have a different functional form since in the immediate vicinity of the TP its asymmetric hopping rules perturb essentially the monolayer particles dynamics.

Next, Eqs.(11) and (12) possess some intrinsic symmetries and hence the number of independent parameters can be reduced. Namely, reversing the field, i.e. changing $\mathbf{E} \to -\mathbf{E}$, leads to the mere replacement of $k(\mathbf{e_1}; t)$ by $k(\mathbf{e_{-1}}; t)$ but does not affect $k(\mathbf{e}_{\boldsymbol{\nu}}; t)$ with $\nu = \pm 2$, which implies that

$$k(\mathbf{e_1}; t)(-\mathbf{E}) = k(\mathbf{e_{-1}}; \mathbf{t})(\mathbf{E}), \text{ and } k(\mathbf{e}_{\boldsymbol{\nu}}; t)(-\mathbf{E}) = k(\mathbf{e}_{\boldsymbol{\nu}}; t)(\mathbf{E}) \text{ for } \nu = \pm 2, \tag{15}$$

Besides, since the transition probabilities in Eq.(2) obey $p_2 = p_{-2}$ one evidently has that

$$k(\mathbf{e_2}; t) = k(\mathbf{e_{-2}}; t), \tag{16}$$

and hence, by symmetry,

$$A_2(t) = A_{-2}(t) \tag{17}$$

which somewhat simplifies Eqs.(11) and (12).

Lastly, we note that despite the fact that using the decoupling scheme in Eq.(10) we effectively close the system of equations on the level of the pair correlations, solution of Eqs.(11) and (12) still poses serious technical difficulties: Namely, these equations are non-linear with respect to the TP velocity, which enters the gradient term on the rhs of the evolution equations for the pair correlation, and does depend itself on the values of the monolayer particles densities in the immediate vicinity of the TP. Below we discuss a solution to this non-linear problem, focusing on the limit $t \to \infty$.

4 Stationary solution of the evolution equations

We turn to the limit $t \to \infty$ and suppose that both the density profiles and stationary velocity of the TP have non-trivial stationary values

$$k(\boldsymbol{\lambda}) \equiv \lim_{t\to\infty} k(\boldsymbol{\lambda}; t), \quad V_{tr} \equiv \lim_{t\to\infty} V_{tr}(t), \quad \text{and} \quad A_\mu \equiv \lim_{t\to\infty} A_\mu(t) \tag{18}$$

Define next the local deviations of $k(\boldsymbol{\lambda})$ from the unperturbed density as

$$h(\boldsymbol{\lambda}) \equiv k(\boldsymbol{\lambda}) - \rho_s \tag{19}$$

Choosing that $h(\mathbf{0}) = 0$, we obtain then the following fundamental system of equations:

$$\tilde{L} h(\boldsymbol{\lambda}) = 0, \tag{20}$$

which holds for all $\boldsymbol{\lambda}$ except for $\boldsymbol{\lambda} = \{\mathbf{0}, \mathbf{e}_{\pm\mathbf{1}}, \mathbf{e}_{\pm\mathbf{2}}\}$, while for the special sites adjacent to the TP, i.e. for $\boldsymbol{\lambda} = \{\mathbf{0}, \mathbf{e}_{\pm 1}, \mathbf{e}_{\pm 2}\}$, one has

$$(\tilde{L} + A_\nu) h(\mathbf{e}_{\boldsymbol{\nu}}) + \rho_s (A_\nu - A_{-\nu}) = 0, \tag{21}$$

Equations (20) and (21) determine the deviation from the unperturbed density ρ_s in the stationary state. Note also that in virtue of the symmetry relations in Eqs.(16) and (17), $h(\mathbf{e}_{\mathbf{2}}) = h(\mathbf{e}_{-\mathbf{2}})$ and $A_2 = A_{-2}$.

Now, our general approach to solution of coupled non-linear Eqs.(4),(20) and (21) is as follows: We first derive a general solution of these equations supposing that V_{tr} is a given parameter, or, in other words, assuming that the coefficients A_ν entering Eqs.(20) and (21) are known. In doing so, we obtain $h(\boldsymbol{\lambda})$ in the parametrized form

$$h(\boldsymbol{\lambda}) = h(\boldsymbol{\lambda}; A_{\pm 1}, A_2). \tag{22}$$

Then, substituting into Eq.(22) particular values $\boldsymbol{\lambda} = \{\mathbf{e}_{\pm 1}, \mathbf{e}_{\pm 2}\}$ and making use of the definition of A_μ in Eq.(14), we find a system of three linear equations with three unknown coefficients of the form

$$A_\nu = 1 + \frac{4\tau^*}{\tau} p_\nu \Big(1 - \rho_s - h(\mathbf{e}_{\boldsymbol{\nu}}; A_{\pm 1}, A_2)\Big), \tag{23}$$

where $\nu = \{\pm 1, 2\}$, which will allow us to define all A_ν (and hence, all $h(\mathbf{e}_{\boldsymbol{\nu}})$). Finally, substituting the results into Eq.(4), which can be written down in terms of A_ν as

$$V_{tr} = \frac{\sigma}{4\tau^*}(A_1 - A_{-1}), \tag{24}$$

we will arrive at a closed-form equation determining implicitly the stationary velocity.

4.1 Generating function for the stationary particle density profiles in the monolayer.

Equations (20) and (21) can be most conveniently solved by introducing the generating function for the density profiles of the form

$$H(w_1, w_2) \equiv \sum_{n_1=-\infty}^{+\infty} \sum_{n_2=-\infty}^{+\infty} h_{n_1,n_2} w_1^{n_1} w_2^{n_2}, \tag{25}$$

where $h_{n_1,n_2} \equiv h(n_1\mathbf{e_1} + n_2\mathbf{e_2})$. Multiplying both sides of Eqs.(20) and (21) by $w_1^{n_1} w_2^{n_2}$, and performing summations over n_1 and n_2 we find that $H(w_1, w_2)$ is given explicitly by

$$H(w_1, w_2) \quad = \quad - K(w_1, w_2) \left\{ A_1 w_1^{-1} + A_{-1} w_1 + A_2(w_2 + w_2^{-1}) - \alpha \right\}^{-1}, \tag{26}$$

where

$$\alpha \equiv \sum_{\nu} A_{\nu} + 4(f + g), \tag{27}$$

and

$$K(w_1, w_2) \equiv \sum_{\nu} A_{\nu} (w_{|\nu|}^{\nu/|\nu|} - 1) h(\mathbf{e}_{\boldsymbol{\nu}}) + \rho_s (A_1 - A_{-1})(w_1 - w_1^{-1}). \tag{28}$$

Equations (26) to (28) determine the generation function for the density profiles exactly, and the latter can be explicitly obtained via standard inversion formulae.

4.2 Integral characteristic of the density profiles

As we have already remarked, the presence of the driven TP induces an inhomogeneous density distribution in the monolayer. One can thus pose a natural question whether equilibrium between adsorption and desorption processes gets shifted due to such a perturbancy, i.e. whether the equilibrium density in the monolayer is different from that given by Eq.(1). The answer is trivially "no" in the case when the particles number is explicitly conserved, but in the general case with arbitrary f and g this is not at all evident: similarly to the behavior in one-dimensional system [21], one expects that also in two-dimensions the density profiles are asymmetric as seen from the stationary moving TP and are characterized by a condensed, "traffic-jam"-like region in front of and a depleted region past the TP. One anticipates then that the desorption events are favored in front of the TP, while the adsorption events are evidently suppressed by the excess density. On the other hand, past the TP desorption is diminished due to the particles depletion while adsorption may proceed more readily due to the same reason. It is thus not at all clear *a priori* whether these two effects can compensate each other exactly, in view of a possible asymmetry of the density profiles, as it happens in the one-dimensional model [21].

For this purpose, we study the behavior of the integral deviation Ω of the density from the equilibrium value ρ_s, i.e.

$$\Omega \equiv \sum_{n_1=-\infty}^{+\infty} \sum_{n_2=-\infty}^{+\infty} h_{n_1,n_2}, \tag{29}$$

which can be computed straightforwardly from Eqs.(26) and (28) by setting both w_1 and w_2 equal to unity. Noticing that $K(w_1 = 1, w_2 = 1) = 0$, and that $A_1+A_{-1}+2A_2-\alpha = -4(f+g)$, i.e. is strictly negative as soon as adsorption/desorption processes are present, we obtain then that Ω is stricly equal to 0. This implies, in turn, that the perturbancy of the density distribution in the monolayer created by the driven TP does not shift the global balance between the adsorption and desorption events. An analogous result has been obtained for the one-dimensional problem in Ref.[21].

4.3 Inversion of the generating function with respect to the "symmetric" coordinate w_2.

Inversion of the generating function with respect to w_2 can be readily performed by expanding $H(w_1, w_2)$ into the series in powers of w_2. To do this, we first rewrite $H(w_1, w_2)$ as

$$H(w_1, w_2) = \alpha^{-1} K(w_1, w_2) \sum_{i=0}^{+\infty} \alpha^{-i} \Big(A_1 w_1^{-1} + A_{-1} w_1 + A_2 (w_2 + w_2^{-1}) \Big)^i. \tag{30}$$

Then, the sum on the right-hand-side of Eq.(30) can be further expanded as

$$\begin{aligned} &\sum_{i=0}^{+\infty} \alpha^{-i} \Big(A_1 w_1^{-1} + A_{-1} w_1 + A_2 (w_2 + w_2^{-1}) \Big)^i \\ = &\sum_{n_2=-\infty}^{+\infty} \sum_{i=0}^{+\infty} \sum_{j=0}^{+\infty} \alpha^{-(i+|n_2|+2j)} \binom{i+|n_2|+2j}{|n_2|+2j} \binom{|n_2|+2j}{j} (A_1 w_1^{-1} + A_{-1} w_1)^i A_2^{|n_2|+2j} w_2^{n_2}, \end{aligned} \tag{31}$$

where $\binom{n}{p}$ stands for the binomial coefficients. Next, gathering the terms with the same power of w_2 on the two sides of Eq. (30) and using Eq. (31), we have that

$$\begin{aligned} \sum_{n_1=-\infty}^{+\infty} h_{n_1,n_2} w_1^{n_1} &= \alpha^{-1} \Big\{ -\sum_{\nu} A_\nu h(\mathbf{e}_{\boldsymbol{\nu}}) + \Big(A_1 h(\mathbf{e_1}) + \rho_s (A_1 - A_{-1}) \Big) w_1 \\ &+ \Big(A_{-1} h(\mathbf{e_{-1}}) - \rho_s (A_1 - A_{-1}) \Big) w_1^{-1} \Big\} F^{(2)}(w_1, |n_2|) \\ &+ \alpha^{-1} A_2 h(\mathbf{e_2}) \Big(F^{(2)}(w_1, |n_2 - 1|) + F^{(2)}(w_1, |n_2 + 1|) \Big), \end{aligned} \tag{32}$$

where

$$F^{(2)}(w_1, n_2) \equiv \sum_{i=0}^{+\infty} \sum_{j=0}^{+\infty} \alpha^{-(i+n_2+2j)} \binom{i+n_2+2j}{n_2+2j} \binom{n_2+2j}{j} A_2^{n_2+2j} (A_1 w_1^{-1} + A_{-1} w_1)^i. \tag{33}$$

As the next step, we resum the series entering $F^{(2)}(w_1, n_2)$. Substituting to Eq.(33) explicit expressions for the binomial coefficients and for the Gamma function, we have

$$\begin{aligned} F^{(2)}(w_1, n_2) &= \left(\frac{A_2}{\alpha}\right)^{n_2} \sum_{j=0}^{+\infty} \Big\{ \left(\frac{A_2}{\alpha}\right)^{2j} \frac{1}{\Gamma(j+1)\Gamma(n_2+j+1)} \\ &\times \int_0^{+\infty} e^{-t} t^{n_2+2j} \sum_{i=0}^{+\infty} \left(\frac{A_1 w_1^{-1} + A_{-1} w_1}{\alpha}\right)^i \frac{t^i}{\Gamma(i+1)} \mathrm{d}t \Big\}, \end{aligned} \tag{34}$$

which yields

$$\begin{aligned} F^{(2)}(w_1, n_2) &= \left(\frac{A_2}{\alpha}\right)^{n_2} \left(1 - \frac{A_1 w_1^{-1} + A_{-1} w_1}{\alpha}\right)^{-(n_2+1)} \\ &\times \sum_{j=0}^{+\infty} \left(\frac{A_2}{\alpha - (A_1 w_1^{-1} + A_{-1} w_1)}\right)^{2j} \binom{n_2+2j}{j}. \end{aligned} \tag{35}$$

Next, using the identity

$$\sum_{j=0}^{+\infty} x^{2j} \binom{n_2+2j}{j} = 2^{n_2} (1-4x^2)^{-1/2} \left(1 + \sqrt{1-4x^2}\right)^{-n_2}, \tag{36}$$

we obtain

$$F^{(2)}(w_1, n_2) = \frac{\alpha y}{A_2 \sqrt{1-4y^2}} \left(\frac{2y}{1+\sqrt{1-4y^2}}\right)^{n_2}, \tag{37}$$

where $y = A_2/(\alpha - (A_1 w_1^{-1} + A_{-1} w_1))$. Eqs.(32) to (37) define the inverted with respect to w_2 generating function.

4.4 Inversion of the generating function with respect to the "asymmetric" coordinate w_1.

Inversion of $H(w_1, w_2)$ with respect to w_1 can be performed along exactly the same lines as we did it in the previous subsection for the variable w_2. That is, by expanding $F^{(2)}(w_1, n_2)$ in Eq.(37) into a series in powers of w_1:

$$\begin{aligned} F^{(2)}(w_1, n_2) = \left(A_{sign(-n_1)}\right)^{|n_1|} \sum_{n_1=-\infty}^{+\infty} \sum_{k=0}^{+\infty} \sum_{j=0}^{+\infty} \Big\{ \alpha^{-(|n_1|+n_2+2j+2k)} \times \\ \times \binom{|n_1|+n_2+2j+2k}{n_2+2j} \binom{n_2+2j}{j} \binom{|n_1|+2k}{k} A_2^{n_2+2j} (A_1 A_{-1})^k w_1^{n_1} \Big\}. \end{aligned} \tag{38}$$

and then identifying the coefficients in this expansion with the analogous coefficients in the expansion in powers of the variable w_1 of

$$h_{n_1,n_2} = \alpha^{-1} \Big\{ \sum_\nu A_\nu h(\mathbf{e}_{\boldsymbol{\nu}}) \nabla_{-\nu} F_{n_1,n_2} - \rho_s (A_1 - A_{-1})(\nabla_1 - \nabla_{-1}) F_{n_1,n_2} \Big\}. \tag{39}$$

with

$$F_{n_1,n_2} \equiv \left(A_{sign(-n_1)}\right)^{|n_1|} \sum_{k=0}^{+\infty}\sum_{j=0}^{+\infty} \Big\{ \alpha^{-(|n_1|+|n_2|+2j+2k)} \times \binom{|n_1|+|n_2|+2j+2k}{|n_2|+2j}\binom{|n_2|+2j}{j}\binom{|n_1|+2k}{k} A_2^{|n_2|+2j}(A_1A_{-1})^k \Big\} \quad (40)$$

Then, using an integral identity

$$\binom{|n_1|+|n_2|+2j+2k}{|n_2|+2j}\binom{|n_2|+2j}{j}\binom{n_1+2k}{k} = \int_0^\infty e^{-t} t^{|n_1|+|n_2|+2k+2j+1} \frac{\mathrm{d}t}{\Gamma(k+1)\Gamma(|n_1|+k+1)\Gamma(j+1)\Gamma(|n_2|+j+1)} \quad (41)$$

we cast F_{n_1,n_2} into the form

$$F_{n_1,n_2} = \left(\frac{A_{-1}}{A_1}\right)^{n_1/2} \int_0^\infty e^{-t} \mathrm{I}_{n_1}\left(2\alpha^{-1}\sqrt{A_1A_{-1}}t\right) \mathrm{I}_{n_2}\left(2\alpha^{-1}A_2 t\right) \mathrm{d}t, \quad (42)$$

where $\mathrm{I}_n(z)$ stands for the modified Bessel, defined as

$$\mathrm{I}_n(z) = \frac{1}{2\pi}\int_{-\pi}^{\pi} d\theta \, \cos(n\theta)\exp(z\cos(\theta)) \quad (43)$$

It is worth-while to mention now that F_{n_1,n_2} has an interesting physical interpretation. To illustrate it, we rewrite the integral involved in Eq.(42) using the definition in Eq.(43) as

$$\int_0^\infty e^{-t} \mathrm{I}_{n_1}\left(2\alpha^{-1}\sqrt{A_1A_{-1}}t\right) \mathrm{I}_{n_2}\left(2\alpha^{-1}A_2 t\right) \mathrm{d}t = \frac{1}{(2\pi)^2}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} \frac{e^{-(k_1n_1+k_2n_2)}}{1-\xi\lambda(\mathbf{k})} \mathrm{d}k_1\mathrm{d}k_2, \quad (44)$$

where

$$\xi = 2\alpha^{-1}(\sqrt{A_1A_{-1}} + A_2), \quad \text{and} \quad \lambda(\mathbf{k}) = \frac{\sqrt{A_1A_{-1}}\cos(k_1) + A_2\cos(k_2)}{\sqrt{A_1A_{-1}} + A_2}. \quad (45)$$

One recognizes then (cf. ref. [28]) that the rhs of Eq. (44) is the generating function

$$P(n_1, n_2; \xi) \equiv \sum_{j=0}^{+\infty} P_j(n_1, n_2)\xi^j, \quad \xi < 1, \quad (46)$$

of $P_j(n_1, n_2)$ - the probability that a particle performing some special type of random walk on the sites of a two-dimensional square lattice and starting at the origin arrives exactly on the j-th step to the site with the lattice vector $n_1\mathbf{e_1} + n_2\mathbf{e_2}$. This special random walk is characterized by $\lambda(\mathbf{k})$ - the structure factor (cf. ref. [28]), which defines a biased random walk with the following elementary jump probabilities

$$\frac{\sqrt{A_1A_{-1}}}{2\left(\sqrt{A_1A_{-1}} + A_2\right)}, \quad \text{in the directions} \quad \pm\,\mathbf{e_1} \text{ of the lattice,} \quad (47)$$

$$\frac{A_2}{2\left(\sqrt{A_1 A_{-1}}+A_2\right)}, \quad \text{in the directions} \quad \pm\,\mathbf{e_2} \ \text{ of the lattice.} \tag{48}$$

Hence, F_{n_1,n_2} can be interpreted as

$$F_{n_1,n_2} = \left(\frac{A_{-1}}{A_1}\right)^{n_1/2} P(n_1, n_2; \xi), \tag{49}$$

and thus can be thought of as the generating function of a two-dimensional biased random walk.

5 Asymptotical behavior of the density profiles at large distances from the tracer particle.

Equations (32) to (37) allow us to deduce an asymptotical behavior of the density profiles in front of and past the stationary moving TP. To do this, note first that these asymptotic forms of $h_{n,0}$ with $n \to \pm\infty$ can be most straightforwardly obtained from the generating function of $h_{n,0}$:

$$N(z) \equiv \sum_{n=-\infty}^{+\infty} h_{n,0} z^n, \tag{50}$$

which we proceed now to calculate. Using Eqs.(32) and (37), we find that

$$\begin{aligned} N(z) \quad = \quad & \frac{z(z-1)\Big(A_1 h(\mathbf{e_1}) + \rho_s(A_1 - A_{-1})\Big) + (1-z)\Big(A_{-1} h(\mathbf{e_{-1}}) - \rho_s(A_1 - A_{-1})\Big)}{A_{-1}\sqrt{(z-z_1)(z-z_2)(z-z_3)(z-z_4)}} \\ & + h(\mathbf{e_2})\left(\sqrt{\frac{(z-z_2)(z-z_3)}{(z-z_1)(z-z_4)}} - 1\right), \end{aligned} \tag{51}$$

where the roots z_i are defined as

$$z_1 = \frac{1}{A_{-1}}\left\{\frac{\alpha}{2} + A_2 - \sqrt{\left(\frac{\alpha}{2}+A_2\right)^2 - A_1 A_{-1}}\right\}, \tag{52}$$

$$z_2 = \frac{1}{A_{-1}}\left\{\frac{\alpha}{2} - A_2 - \sqrt{\left(\frac{\alpha}{2}-A_2\right)^2 - A_1 A_{-1}}\right\}, \tag{53}$$

$$z_3 = \frac{1}{A_{-1}}\left\{\frac{\alpha}{2} - A_2 + \sqrt{\left(\frac{\alpha}{2}-A_2\right)^2 - A_1 A_{-1}}\right\}, \tag{54}$$

$$z_4 = \frac{1}{A_{-1}}\left\{\frac{\alpha}{2} + A_2 + \sqrt{\left(\frac{\alpha}{2}+A_2\right)^2 - A_1 A_{-1}}\right\}, \tag{55}$$

which obey

$$0 < z_1 \leq z_2 \leq 1 < z_3 < z_4. \tag{56}$$

In consequence, $N(z)$ is a holomorphic function in the annular region of inner radius z_2 and the outer radius z_3. As explained in Appendix B, the behavior of $h_{n,0}$ when $n \to +\infty$ (resp. $n \to -\infty$) is controled by z_3 (resp. z_2^{-1}).

5.1 Asymptotics of density profiles at large distances in front of the tracer particle.

The asymptotical behavior of the density profiles in front of the TP is supported by the behavior of the generating function $N(z)$ in the vincinity of z_3 (see Apendix B for more details). We find then that for $n \to +\infty$ the density obeys

$$h_{n,0} \sim \frac{z_3(z_3-1)\Big(A_1 h_{\mathbf{e_1}} + \rho_s(A_1 - A_{-1})\Big) + (1-z_3)\Big(A_{-1} h_{\mathbf{e_{-1}}} - \rho_s(A_1 - A_{-1})\Big)}{A_{-1}\sqrt{(z_3-z_1)(z_3-z_2)(z_4-z_3)}} \frac{1}{\sqrt{\pi n}} \frac{1}{z_3^n}, \tag{57}$$

which means that in front of the TP the deviation $h_{n,0}$ decays exponentially with the distance,

$$h_{n,0} \sim K_+ \frac{\exp\Big(-n/\lambda_+\Big)}{n^{1/2}}, \tag{58}$$

where the decay amplitude obeys

$$\begin{aligned} K_+ &= \frac{(z_3-1)\Big(A_1 h(\mathbf{e_1}) + \rho_s(A_1 - A_{-1})\Big) + (1/z_3 - 1)\Big(A_{-1} h(\mathbf{e_{-1}}) - \rho_s(A_1 - A_{-1})\Big)}{\sqrt{8\pi A_2}} \\ &\times \left\{ \left(\frac{\alpha/2 - A_2}{A_{-1}}\right)^2 - \frac{A_1}{A_{-1}} \right\}^{-1/4}, \end{aligned} \tag{59}$$

while the characteristic decay length is given by

$$\lambda_+ = 1/\ln(z_3). \tag{60}$$

Note that here λ_+ is finite for any values of the system parameters.

5.2 Asymptotics of density profiles at large distances past the tracer particle.

We first note that one of the roots of the generating function, namely z_2, gets equal to unity when both f and g are strictly equal to zero, which results in the exact cancellation of the multiplier $\sqrt{1-z}$ both in the nominator and the denominator in the first term on the rhs of Eq.(51). This signifies that in the limit when exchanges with the reservoir are

forbidden, the singular behavior of the generating function at the vicinity of z_2 is essentially different compared to the case when both f and g are greater than zero. Consequently, one has to consider separately the behavior in the case of non-conserved particles number, when exchanges with the reservoir persist, and the case when both f and g are equal to zero while their ratio is kept fixed.

A. Non-conserved particles number. In case when both f and g have non-zero values, we find (cf. Appendix B) that when $n \to +\infty$ the density deviation follows

$$h_{-n,0} \sim K_{-} \frac{\exp\Big(-n/\lambda_{-}\Big)}{n^{1/2}}, \tag{61}$$

where the amplitude is given by

$$\begin{aligned} K_{-} \quad &= \quad \frac{(z_2-1)\Big(A_1 h_{\mathbf{e_1}} + \rho_s(A_1 - A_{-1})\Big) + (1/z_2 - 1)\Big(A_{-1} h_{\mathbf{e_{-1}}} - \rho_s(A_1 - A_{-1})\Big)}{\sqrt{8\pi A_2}} \\ &\times \left\{ \left(\frac{\alpha/2 - A_2}{A_1}\right)^2 - \frac{A_{-1}}{A_1} \right\}^{-1/4}, \end{aligned} \tag{62}$$

and the characteristic decay length obeys

$$\lambda_{-} = -1/\ln(z_2). \tag{63}$$

Consequently, in the case when particles exchanges with the reservoir are permitted, the density deviation from the equilibrium value ρ_s decays exponentially with the distance from the TP. Note that the decay lenghts satisfy the inequality $\lambda_{-} > \lambda_{+}$, which means that the correlations between the tracer and particles of the monolayer are always stronger past than in front of the TP. Note also that λ_{-} diverges when both f and g tend to zero, which signals, as we have already remarked, that the decay of $h_{n,0}$ may proceed differently in this case, compared to the exponential dependence in Eq.(61).

B. Conserved particles number. Suppose now that both f and g are equal to zero, while their ratio is fixed and obeys $f/g = \rho_s/(1-\rho_s)$. As we have already remarked, this situation corresponds to the customary model of a two-dimensional hard-core lattice gas without exchanges with a reservoir. For this situation, we find (see Appendix B for more details), that when $n \to +\infty$ the deviation of the particle density from the equilibrium value ρ_s obeys

$$h_{-n,0} = -\frac{K'_{-}}{n^{3/2}} \left(1 + \frac{3}{8n} + \mathcal{O}\Big(\frac{1}{n^2}\Big)\right), \tag{64}$$

where the decay amplitude K'_{-} is given by

$$K'_{-} = \frac{1}{4\sqrt{\pi}}\Bigg\{(A_1 h(\mathbf{e_1}) - A_{-1}h(\mathbf{e_{-1}}) + 2\rho_s(A_1 - A_{-1}))\sqrt{\frac{1}{A_2(A_1 - A_{-1})}} + h(\mathbf{e_2})\sqrt{\frac{A_1 - A_{-1}}{A_2}}\Bigg\} \tag{65}$$

Remarkably enough, in this case the correlations between the TP position and the particles distribution vanish *algebraically* slow with the distance! This implies, in turn, that in the conserved particles number case, the mixing of the monolayer is not efficient enough to prevent the appearence of the quasi-long-range order and the medium "remembers" the passage of the TP on a long time and space scale, which signifies very strong memory effects. We note also that the algebraic decay of correlations in this model has been predicted earlier in Ref.[20]. However, the decay exponent has been erroneously suggested to be equal to 1/2, as opposed to the value 3/2 given by Eq.(64). As well, the amplitude K'_{-} happens to have a different sign, compared to that obtained in Ref.[20], which invalidates the conclusion that the overall relaxation to the equilibrium value ρ_s might show a non-monotoneous behavior as a function of the distance past the TP.

6 Formal expression for the density profiles and general force-velocity relation.

The Eqs.(39) and (42) display h_{n_1,n_2} as a function of the coefficients A_ν that remain to be determined. As a matter of fact, these coefficients depend themselves on the local densities in the immediate vicintiy of the tracer, i.e. on $h(\mathbf{e}_\nu)$. This implies that we have to determine them from Eqs.(39) and (42) in a self-consistent way.

Setting in Eq.(39) $\boldsymbol{\lambda} = \mathbf{e}_{\boldsymbol{\nu}}$, where $\nu = \{\pm 1, 2\}$, results in the following system of equations

$$\tilde{C}\tilde{h} = \rho_s(A_1 - A_{-1})\tilde{F}, \tag{66}$$

where

$$\tilde{h} = \begin{pmatrix} h(\mathbf{e_1}) \\ h(\mathbf{e_{-1}}) \\ h(\mathbf{e_2}) \end{pmatrix}, \quad \tilde{F} = \begin{pmatrix} (\nabla_1 - \nabla_{-1})F_{\mathbf{e_1}} \\ (\nabla_1 - \nabla_{-1})F_{\mathbf{e_{-1}}} \\ (\nabla_1 - \nabla_{-1})F_{\mathbf{e_2}} \end{pmatrix}, \tag{67}$$

and

$$\tilde{C} = \begin{pmatrix} A_1\nabla_{-1}F_{\mathbf{e_1}} - \alpha & A_{-1}\nabla_1 F_{\mathbf{e_1}} & A_2\nabla_{-2}F_{\mathbf{e_1}} \\ A_1\nabla_{-1}F_{\mathbf{e_{-1}}} & A_{-1}\nabla_1 F_{\mathbf{e_{-1}}} - \alpha & A_2\nabla_{-2}F_{\mathbf{e_{-1}}} \\ A_1\nabla_{-1}F_{\mathbf{e_2}} & A_{-1}\nabla_1 F_{\mathbf{e_2}} & A_2\nabla_{-2}F_{\mathbf{e_2}} - \alpha \end{pmatrix}. \tag{68}$$

The solutions of this 3×3 linear system are

$$\forall \nu = \{\pm 1, 2\}, \quad h(\mathbf{e}_{\boldsymbol{\nu}}) = \rho_s (A_1 - A_{-1}) \frac{\det \tilde{C}_\nu}{\det \tilde{C}}, \tag{69}$$

where $\tilde{C}_\nu$ stands for the matrix obtained from $\tilde{C}$ by replacing the ν-th column by the column-vector $\tilde{F}$. The substitution of expression in Eq.(69) into the definition of the coefficients A_ν, (14), results in the following system of three equations

$$\forall \nu = \{\pm 1, 2\}, \quad A_\nu = 1 + \frac{4\tau^*}{\tau} p_\nu \left\{ 1 - \rho_s - \rho_s (A_1 - A_{-1}) \frac{\det \tilde{C}_\nu}{\det \tilde{C}} \right\}, \tag{70}$$

which implicitly determines all unknown coefficients A_ν and hence, the local deviations $h(\mathbf{e}_{\boldsymbol{\nu}})$, defined as

$$h(\mathbf{e}_{\boldsymbol{\nu}}) = (1 - \rho_s) + \frac{\tau}{4\tau^* p_\nu} (1 - A_\nu). \tag{71}$$

Finally, for every lattice point, except for the origin occupied by the TP, the density profiles are given by Eq.(39), where F_{n_1,n_2} is defined by Eq. (42).

Next, substituting Eq.(69) to Eqs.(4) and (19), we find that the TP terminal velocity obeys

$$V_{tr} = \frac{\sigma}{\tau} \Big\{ (p_1 - p_{-1})(1 - \rho_s) - \rho_s (A_1 - A_{-1}) \frac{p_1 \det \tilde{C}_1 - p_{-1} \det \tilde{C}_{-1}}{\det \tilde{C}} \Big\}, \tag{72}$$

which can be rewritten, taking into account Eq.(24), as

$$V_{tr} = \frac{\sigma}{\tau} (p_1 - p_{-1})(1 - \rho_s) \Big\{ 1 + \rho_s \frac{4\tau^*}{\tau} \frac{p_1 \det \tilde{C}_1 - p_{-1} \det \tilde{C}_{-1}}{\det \tilde{C}} \Big\}^{-1}. \tag{73}$$

This last equation represents the desired general force-velocity relation for the system under study, which is valid for arbitrary magnitude of external bias and arbitrary values of other system's parameters. In the general form as it is, Eq.(73) is apparently not very useful, but may describe physically interesting behavior in two particular limits - the case of infinitely strong bias, when the TP performs totally directed random walk, which is appropriate to motion of the AFM tip, and the case when the bias is vanishingly small. While the former case has been studied in detail in Ref.[22], the results for the latter case have been only briefly outlined in Ref.[23]. In the next Section we present detailed derivation of the limiting form of the force-velocity relation in case of a vanishingly small external force.

7 Vanishingly small external bias.

We turn now to the limit $\beta E \ll 1$, in which case the problem simplifies considerably and allows to obtain explicit results for the local densities in the immediate vicinity of the TP and consequently, the TP terminal velocity and diffusivity.

7.1 Local particle density in the vicinity of the tracer in the limit $\beta E \ll 1$.

Setting $\boldsymbol{\lambda} = \mathbf{e_1}$ and $\boldsymbol{\lambda} = \mathbf{e_{-1}}$ in Eq.(39), we obtain equations obeyed by $h(\mathbf{e_{\pm 1}})$:

$$\alpha h(\mathbf{e_1}) = \sum_\nu A_\nu h(\mathbf{e_\nu}) \nabla_{-\nu} F_{\mathbf{e_1}} - \rho_s (A_1 - A_{-1})(\nabla_1 - \nabla_{-1}) F_{\mathbf{e_1}}, \tag{74}$$

and

$$\alpha h(\mathbf{e_{-1}}) = \sum_\nu A_\nu h(\mathbf{e_\nu}) \nabla_{-\nu} F_{\mathbf{e_{-1}}} - \rho_s (A_1 - A_{-1})(\nabla_1 - \nabla_{-1}) F_{\mathbf{e_{-1}}}. \tag{75}$$

Consequently, the discontinuity in the density profile is given by:

$$\begin{aligned} \delta h = \Big(h(\mathbf{e_1}) - h(\mathbf{e_{-1}}) \Big) \quad &= \quad \alpha^{-1} \sum_\nu A_\nu h(\mathbf{e_\nu}) \nabla_{-\nu} (F_{\mathbf{e_1}} - F_{\mathbf{e_{-1}}}) - \\ &- \quad \rho_s (A_1 - A_{-1})(\nabla_1 - \nabla_{-1})(F_{\mathbf{e_1}} - F_{\mathbf{e_{-1}}}) \end{aligned} \tag{76}$$

Our aim is now to compute the leading order contribution to δh in the limit $\beta E \ll 1$. To this purpose, we first expand p_ν, $h(\mathbf{e_{\pm 1}})$ and A_ν in the Taylor series in powers of E retaining only linear with the field terms:

$$p_\nu = \frac{1}{4}\Big(1 + \frac{\beta\sigma}{2} (\mathbf{E} \cdot \mathbf{e_\nu}) + \mathcal{O}(E^2) \Big), \tag{77}$$

$$h_{\mathbf{e_1}} \propto E \quad \text{and} \quad h_{\mathbf{e_{-1}}} = -h_{\mathbf{e_1}} + \mathcal{O}(E^2), \tag{78}$$

$$h_{\mathbf{e_{\pm 2}}} = \mathcal{O}(E^2), \tag{79}$$

and consequently,

$$A_\nu = 1 + \frac{\tau^*}{\tau}(1 - \rho_s) + \frac{\tau^*}{\tau}\Big((1 - \rho_s) \frac{\beta\sigma}{2} (\mathbf{E} \cdot \mathbf{e_\nu}) - h(\mathbf{e_\nu}) \Big) + \mathcal{O}(E^2), \tag{80}$$

where we have made use of the symmetry relations given by Eq. (15). Using these equations, we can expand the terms in the right-hand-side of Eq.(76) as:

$$\begin{aligned} \sum_\nu A_\nu h(\mathbf{e_\nu}) \nabla_{-\nu} (F_{\mathbf{e_1}} \quad &- \quad F_{\mathbf{e_{-1}}}) = \sum_\nu A_\nu h(\mathbf{e_\nu}) \nabla_{-\nu} (F_{\mathbf{e_1}}(E=0) - F_{\mathbf{e_{-1}}}(E=0)) + \mathcal{O}(E^2) \\ &= \quad h_{\mathbf{e_1}} (A_1 \nabla_{-1} - A_{-1} \nabla_1)(F_{\mathbf{e_1}}(E=0) - F_{\mathbf{e_{-1}}}(E=0)) + \mathcal{O}(E^2) \\ &= \quad \frac{2h(\mathbf{e_1})}{\mathcal{L}(2A_0/\alpha_0)} \left(1 + \frac{\tau^*}{\tau}(1 - \rho_s) \right) + \mathcal{O}(E^2), \end{aligned} \tag{81}$$

where

$$\mathcal{L}(x) \equiv \left\{ \int_0^\infty e^{-t} \Big((\mathrm{I}_0(xt) - \mathrm{I}_2(xt)) \mathrm{I}_0(xt) \mathrm{d}t \right\}^{-1}, \tag{82}$$

$$A_0 \equiv A_\nu(E=0) = 1 + \frac{\tau^*}{\tau}(1-\rho_s), \tag{83}$$

and $\alpha_0 \equiv \alpha(E=0)$. Next, we have that

$$\begin{aligned} \rho_s(A_1 - A_{-1})(\nabla_1 - \nabla_{-1})(F_{\mathbf{e_1}} &- F_{\mathbf{e_{-1}}}) = \\ &- \frac{2\rho_s}{\mathcal{L}(2A_0/\alpha_0)}\frac{\tau^*}{\tau}\left((1-\rho_s)\beta\sigma E - 2h(\mathbf{e_1})\right) + \mathcal{O}(E^2) \end{aligned} \tag{84}$$

Substituting Eqs.(81) and (84) into Eq.(76), we obtain

$$\begin{aligned} h(\mathbf{e_1}) &= \frac{h(\mathbf{e_1})}{\alpha_0\mathcal{L}(2A_0/\alpha_0)}\left(1 + \frac{\tau^*}{\tau}(1-\rho_s)\right) + \\ &+ \frac{\rho_s}{\alpha_0\mathcal{L}(2A_0/\alpha_0)}\frac{\tau^*}{\tau}\left((1-\rho_s)\beta\sigma E - 2h(\mathbf{e_1})\right) + \mathcal{O}(E^2), \end{aligned} \tag{85}$$

which leads to the desired result for the deviation of the local density just in front of the TP from the equilibrium value ρ_s. The leading order contribution to $h(\mathbf{e_1})$ with respect to the field is thus given explicitly by

$$h(\mathbf{e_1}) = \rho_s(1-\rho_s)\beta\sigma E\frac{\tau^*}{\tau}\left\{\alpha_0\mathcal{L}(2A_0/\alpha_0) - 1 + (3\rho_s - 1)\frac{\tau^*}{\tau}\right\}^{-1} + \mathcal{O}(E^2). \tag{86}$$

Hence, the discontinuity δh in the monolayer particles density in the immediate vicinity of the TP equals, by virtue of Eq.(78), twice the expression on the right-hand-side of Eq.(86).

7.2 Friction coefficient and the Stokes formula.

Expanding the TP velocity V_{tr} in the Taylor series in powers of E and again, retaining only linear with E terms, we get

$$V_{tr} = \frac{\beta\sigma^2}{4\tau}(1-\rho_s)E - \frac{\sigma}{4\tau}\Big(h(\mathbf{e_1}) - h(\mathbf{e_{-1}})\Big) + \mathcal{O}(E^2) \tag{87}$$

Next, making use of Eqs.(78) and (86), we arrive at the following explicit result

$$V_{tr} \sim \frac{\beta\sigma^2}{4\tau}(1-\rho_s)E\left\{1 - \frac{2\rho_s\tau^*}{\tau}\frac{1}{\alpha_0\mathcal{L}(2A_0/\alpha_0) - 1 + (3\rho_s-1)\tau^*/\tau}\right\}, \tag{88}$$

which signifies that in the limit of a vanishingly small external bias the frictional force exerted on the TP by the monolayer particles is viscous. Note also that Eq.(88) is quite similar to the well-known Stokes formula, i.e. $V_{tr} \sim E/\zeta$, and in our case the friction coefficient ζ is given explicitly by

$$\zeta = \frac{4\tau}{\beta\sigma^2(1-\rho_s)}\left\{1 + \frac{\tau^*}{\tau}\frac{2\rho_s}{\alpha_0\mathcal{L}(2A_0/\alpha_0) - A_0}\right\}. \tag{89}$$

Note now that the friction coefficient ζ is the sum of two contributions. The first one,

$$\zeta_{cm} \equiv \frac{4\tau}{\beta\sigma^2(1-\rho_s)}, \tag{90}$$

is a typical mean-field-type result and corresponds to a perfectly homogeneous monolayer (see discussion following Eq.(6)). The second one,

$$\zeta_{coop} = \frac{4\tau^*}{\beta\sigma^2(1-\rho_s)} \frac{2\rho_s}{\alpha_0\mathcal{L}_2(2A_0/\alpha_0) - A_0}, \tag{91}$$

has, however, a more complicated origin. Namely, it is associated with the cooperative behavior emerging in the monolayer - dehomogenization of the particle distribution in the adsorbed monolayer due to the presence of a driven impurity (the TP) and formation of stationary density profiles, whose characteristic properties depend on the velocity V_{tr}.

7.3 Diffusion coefficient and Einstein relation

Lastly, assuming *a priori* that the Einstein relation holds for the system under study, we estimate the TP diffusion coefficient D_{tr} as

$$D_{tr} = \beta^{-1}\zeta^{-1} = \frac{\sigma^2}{4\tau}(1-\rho_s)\left\{1 - \frac{2\rho_s\tau^*}{\tau}\frac{1}{\alpha_0\mathcal{L}(2A_0/\alpha_0) - 1 + (3\rho_s - 1)\tau^*/\tau}\right\}. \tag{92}$$

It seems now interesting to compare our general result in Eq.(92) against the classical result of Nakazato and Kitahara [16], which describes TP diffusion coefficient in a two-dimensional lattice gas with conserved particles number. Setting f and g equal to zero, while assuming that their ratio has a fixed value, $f/g = \rho_s/(1-\rho_s)$, we have then that

$$D_{tr} \to \hat{D}_{tr} = \frac{\sigma^2}{4\tau}(1-\rho_s)\left\{1 - \frac{2\rho_s\tau^*}{\tau}\frac{1}{4A_0\mathcal{L}(1/2) - 1 + (3\rho_s - 1)\tau^*/\tau}\right\}. \tag{93}$$

Noticing next that

$$\frac{1}{\mathcal{L}(1/2)} = \lim_{\xi\to 1^-}\Big(P(0,0;\xi) - P(2,0;\xi)\Big), \tag{94}$$

where $P(n_1, n_2; \xi)$ has been defined by Eq.(46), and using the fact that [31]

$$\lim_{\xi\to 1^-}\Big(P(0,0;\xi) - P(2,0;\xi)\Big) = 4 - \frac{8}{\pi}, \tag{95}$$

we find that the right-hand-side of Eq.(93) attains the form

$$\hat{D}_{tr} = \frac{\sigma^2}{4\tau}(1-\rho_s)\left\{1 - \frac{2\rho_s\tau^*}{\tau}\frac{1 - 2/\pi}{1 + (1-\rho_s)\tau^*/\tau - (1-2/\pi)(1 + (1-3\rho_s)\tau^*/\tau)}\right\}, \tag{96}$$

which expression is exactly the same as the one obtained earlier in Refs.[16] and [17] within the framework of different, compared to our approach, analytical techniques. The result in Eq.(96) is known to be exact in the limits $\rho_s \ll 1$ and $\rho_s \sim 1$, and serves as a very good approximation for the self-diffusion coefficient in hard-core lattice gases of arbitrary density [15].

8 Conclusion

To conclude, we have studied analytically the intrinsic frictional properties of 2D adsorbed monolayers, composed of mobile hard-core particles undergoing continuous exchanged with the vapor. Our analytical approach has been based on the master equation, describing the time evolution of the system, which has allowed us to evaluate a system of coupled dynamical equations for the tracer particle velocity and a hierarchy of correlation functions. To solve these coupled equations, we have invoked an approximate closure scheme based on the decomposition of the third-order correlation functions into a product of pairwise correlations, which has been first introduced in Ref.[24] for a related model of a driven tracer particle dynamics in a one-dimensional lattice gas with conserved particles number. Within the framework of this approximation, we have derived a system of coupled, discrete-space equations describing evolution of the density profiles in the adsorbed monolayer, as seen from the moving tracer, and its velocity V_{tr}. We have shown that the density profile around the tracer is strongly inhomogeneous: the local density of the adsorbed particles in front of the tracer is higher than the average and approaches the average value as an exponential function of the distance from the tracer. On the other hand, past the tracer the local density is always lower than the average, and depending on whether the number of particles is explicitly conserved or not, the local density past the tracer may tend to the average value either as an exponential or even as an *algebraic* function of the distance. The latter reveals especially strong memory effects and strong correlations between the particle distribution in the environment and the carrier position. Next, we have derived a general force-velocity relation, which defines the terminal velocity of the tracer particle for arbitrary applied fields and arbitrary values of other system parameters. We have demonstrated next that in the limit of a vanishingly small external bias this relation attains a simple, but physically meaningful form of the Stokes formula, which signifies that in this limit the frictional force exerted on the tracer by the adsorbed monolayer particles is viscous. Corresponding friction coefficient has been also explicitly determined. In addition, we estimated the self-diffusion coefficient of the tracer in the absence of the field and showed that it reduces to the well-know result of Refs.[16] and [17] in the limit when the particles number is conserved.

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Figure Captions.

Fig.1. Adsorbed monolayer in contact with a vapor. Grey spheres denote the monolayer (vapor) particles; the grey sphere with an arrow stands for the driven tracer particle.

Fig.2. Singular points z_i of the generating function $N(z)$ and the integration contour (thick line).

Fig.3. Piecewise contour $\mathcal{H}(n)$ of the Hankel type.

Appendix A

In this Appendix we present the details of calculation of different contributions to the time evolution of the pair correlation function $k(\boldsymbol{\lambda};t)$ (cf. Eq.(7)), associated with four lines in the master equation (3).

Consider first the contribution associated with the hopping motion of the adsorbed particles:

$$
\begin{aligned}
C_1(\boldsymbol{\lambda}) &\equiv \sum_{\mathbf{R_{tr}},\eta}\sum_{\mu=1,2}\sum_{\mathbf{r}\neq\mathbf{R_{tr}}-\mathbf{e}_{\boldsymbol{\mu}},\mathbf{R_{tr}}} \eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})\left\{P(\mathbf{R_{tr}},\eta^{\mathbf{r},\mu};t)-P(\mathbf{R_{tr}},\eta;t)\right\} \\
&= \sum_{\mathbf{R_{tr}},\eta}\sum_{\mu=1,2}\sum_{\mathbf{r}\neq\mathbf{R_{tr}}-\mathbf{e}_{\boldsymbol{\mu}},\mathbf{R_{tr}}} \left\{\eta^{\mathbf{r},\mu}(\mathbf{R_{tr}}+\boldsymbol{\lambda})-\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})\right\}P(\mathbf{R_{tr}},\eta;t).
\end{aligned}
\tag{97}
$$

Now, one has then to distinguish between two possible situations:

(a) when $\boldsymbol{\lambda}\neq\mathbf{e}_{\boldsymbol{\nu}}$, one has

$$
\begin{aligned}
&\sum_{\mu=1,2}\sum_{\mathbf{r}\neq\mathbf{R_{tr}}-\mathbf{e}_{\boldsymbol{\mu}},\mathbf{R_{tr}}} \left\{\eta^{\mathbf{r},\mu}(\mathbf{R_{tr}}+\boldsymbol{\lambda})-\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})\right\}P(\mathbf{R_{tr}},\eta;t) \\
&= \sum_{\mu=1,2}(\nabla_\mu+\nabla_{-\mu})\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda}) \\
&= \sum_{\mu}\nabla_\mu\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda}),
\end{aligned}
\tag{98}
$$

which yields, eventually,

$$
C_1(\boldsymbol{\lambda}) = \sum_\mu \nabla_\mu k(\boldsymbol{\lambda};t).
\tag{99}
$$

(b) when $\boldsymbol{\lambda}=\mathbf{e}_{\boldsymbol{\nu}}$, i.e. at the sites adjacent to the tracer, we find

$$
\begin{aligned}
&\sum_{\mu=1,2}\sum_{\mathbf{r}\neq\mathbf{R_{tr}}-\mathbf{e}_{\boldsymbol{\mu}},\mathbf{R_{tr}}} \left\{\eta^{\mathbf{r},\mu}(\mathbf{R_{tr}}+\mathbf{e}_{\boldsymbol{\nu}})-\eta(\mathbf{R_{tr}}+\mathbf{e}_{\boldsymbol{\nu}})\right\}P(\mathbf{R_{tr}},\eta;t) \\
&= (\sum_{\mu}\nabla_\mu-\nabla_{-\nu})\eta(\mathbf{R_{tr}}+\mathbf{e}_{\boldsymbol{\nu}}),
\end{aligned}
\tag{100}
$$

which implies that

$$
C_1(\mathbf{e}_{\boldsymbol{\nu}}) = (\sum_\mu \nabla_\mu-\nabla_{-\nu})k(\mathbf{e}_{\boldsymbol{\nu}};t).
\tag{101}
$$

Finally, using the Kroneker-delta $\delta_{\boldsymbol{\lambda},\mathbf{e}_{\boldsymbol{\mu}}}$, we can generalize both results for arbitrary $\boldsymbol{\lambda}$:

$$
C_1(\boldsymbol{\lambda}) = (\sum_\mu \nabla_\mu-\delta_{\boldsymbol{\lambda},\mathbf{e}_{\boldsymbol{\mu}}}\nabla_{-\mu})k(\boldsymbol{\lambda};t).
\tag{102}
$$

Next, we turn to the contibution stemming out of random biased hopping motion of the tracer particle. This reads

$$\begin{aligned}
C_2(\boldsymbol{\lambda}) &\equiv \frac{4\tau^*}{\tau}\sum_{\mathbf{R_{tr}},\eta}\sum_{\mu} p_\mu \eta(\mathbf{R}_{tr}+\mathbf{e}_{\boldsymbol{\nu}})\{(1-\eta(\mathbf{R_{tr}}))\,P(\mathbf{R_{tr}}-\mathbf{e}_{\boldsymbol{\mu}},\eta;t) \\
&\qquad -(1-\eta(\mathbf{R_{tr}}+\mathbf{e}_{\boldsymbol{\mu}}))\,P(\mathbf{R_{tr}},\eta;t)\} \\
&= \frac{4\tau^*}{\tau}\sum_{\mathbf{R_{tr}},\eta}\sum_{\mu} p_\mu(1-\eta(\mathbf{R_{tr}}+\mathbf{e}_\mu))\,\{\eta(\mathbf{R}_{tr}+\mathbf{e}_{\boldsymbol{\nu}}+\mathbf{e}_{\boldsymbol{\mu}})-\eta(\mathbf{R}_{tr}+\mathbf{e}_{\boldsymbol{\nu}})\}\,P(\mathbf{R_{tr}},\eta;t) \\
&= \frac{4\tau^*}{\tau}\sum_{\mathbf{R_{tr}},\eta}\sum_{\mu} p_\mu\left(1-\eta(\mathbf{R_{tr}}+\mathbf{e}_{\boldsymbol{\mu}})\right)\nabla_\mu\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})P(\mathbf{R_{tr}},\eta;t), \qquad (103)
\end{aligned}$$

Further on, we consider the contribution associated with desorption of the monolayer particles:

$$\begin{aligned}
C_3(\boldsymbol{\lambda}) &\equiv 4g\sum_{\mathbf{R_{tr}}}\sum_{\mathbf{r}\neq\mathbf{R_{tr}}}\sum_{\eta}\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})\,\{(1-\eta(\mathbf{r}))\,P(\mathbf{R_{tr}},\hat{\eta}^{\mathbf{r}};t)-\eta(\mathbf{r})P(\mathbf{R_{tr}},\eta;t)\} \\
&= 4g\sum_{\mathbf{R_{tr}}}\sum_{\mathbf{r}\neq\mathbf{R_{tr}}}\sum_{\eta}\eta(\mathbf{r})\{\hat{\eta}^{\mathbf{r}}(\mathbf{R_{tr}}+\boldsymbol{\lambda})-\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})\}P(\mathbf{R_{tr}},\eta;t) \\
&= 4g\sum_{\mathbf{R_{tr}}}\sum_{\eta}\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})(1-2\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda}))P(\mathbf{R_{tr}},\eta;t) \qquad (104)
\end{aligned}$$

Taking into account that $\eta(\mathbf{R})$ assumes only two values - 0 and 1, and hence, that $\eta^2(\mathbf{R}) = \eta(\mathbf{R})$, we have

$$C_3(\boldsymbol{\lambda}) = -4gk(\boldsymbol{\lambda};t). \qquad (105)$$

Lastly, the contribution due to adsorption of the particles from the vapor phase onto the lattice reads

$$\begin{aligned}
C_4(\boldsymbol{\lambda}) &\equiv 4f\sum_{\mathbf{R_{tr}},\eta}\sum_{\mathbf{r}\neq\mathbf{R_{tr}}}\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})\,\{\eta(\mathbf{r})P(\mathbf{R_{tr}},\hat{\eta}^{\mathbf{r}};t)-(1-\eta(\mathbf{r}))P(\mathbf{R_{tr}},\eta;t)\} \\
&= 4f\sum_{\mathbf{R_{tr}},\eta}\sum_{\mathbf{r}\neq\mathbf{R_{tr}}}(1-\eta(\mathbf{r}))\{\hat{\eta}^{\mathbf{r}}(\mathbf{R_{tr}}+\boldsymbol{\lambda})-\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda})\}P(\mathbf{R_{tr}},\eta;t) \\
&= 4f\sum_{\mathbf{R_{tr}},\eta}(1-\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda}))(1-2\eta(\mathbf{R_{tr}}+\boldsymbol{\lambda}))P(\mathbf{R_{tr}},\eta;t) \qquad (106)
\end{aligned}$$

Again, since $\eta^2(\mathbf{R}) = \eta(\mathbf{R})$, the latter equation reduces to

$$C_4(\boldsymbol{\lambda}) = -4fk(\boldsymbol{\lambda};t) + 4f. \qquad (107)$$

Summing up all four contributions, we arrive at the evolution equation in Eq.(7).

Appendix B

We present here a derivation of the asymptotic behavior of the density profiles at large separations from the stationary moving tracer. The generating function $N(z)$, defined by Eq.(51), has the roots z_i satisfy Eqs.(52) to (55). Introducing the cuts depicted on Fig.2, one observes that $N(z)$ is an analytic function in the annular region of inner radius z_2 and the outer radius z_3. In consequence, density deviations $h_{n,0}$ from the equilibrium value ρ_s at positions $\boldsymbol{\lambda} = \boldsymbol{e}_1 n$ are given by the Cauchy formula

$$h_{n,0} = \frac{1}{2i\pi} \oint \frac{N(z)}{z^{n+1}} \mathrm{d}z, \tag{108}$$

where the contour of the integration is a positively oriented circle, centered around O, of radius R such that $z_2 < R < z_3$ (see Fig.2).

I. Asymptotic behavior of $h_{n,0}$ in the limit $n \to +\infty$. Following closely the reasonings of Flajolet et al. [32], we notice that the asymptotical behavior of $h_{n,0}$ in the limit $n \to +\infty$ is supported by the behavior of the generating function $N(z)$ in the vicinity of $z = z_i$, where z_i is the first root encountered when one tries to deforme the contour in the integral (108) by increasing its radius. In our case, the relevant root is z_3. Now, we have to choose a contour of integration that comes close enough to "capture" the behavior of $N(z)$ in the vicinity of this leading singularity. Let us formalize this idea.

We begin by expanding $N(z)$ in the vicinity of z_3, which gives

$$N(z) = C(z_3 - z)^{-1/2} - h(\mathbf{e_2}) + \mathcal{O}(\sqrt{z_3 - z}), \tag{109}$$

where C is a constant defined by

$$\begin{aligned} C \equiv & \frac{z_3(z_3-1)\Big(A_1 h(\mathbf{e_1}) + \rho_s(A_1 - A_{-1})\Big) + (1-z_3)\Big(A_{-1} h(\mathbf{e_{-1}}) - \rho_s(A_1 - A_{-1})\Big)}{\sqrt{8A_2 z_3}} \\ \times & \left\{ \left(\frac{\alpha/2 - A_2}{A_{-1}} \right)^2 - \frac{A_1}{A_{-1}} \right\}^{-1/4}. \end{aligned} \tag{110}$$

Then, Eq.(108) attains the form

$$h_{n,0} = \frac{C}{2i\pi} \oint \frac{(z_3 - z)^{-1/2}}{z^{n+1}} \mathrm{d}z - \underbrace{\frac{h(\mathbf{e_2})}{2i\pi} \oint \frac{1}{z^{n+1}} \mathrm{d}z}_{=0} + \frac{1}{2i\pi} \oint \frac{\mathcal{O}(\sqrt{z_3 - z})}{z^{n+1}} \mathrm{d}z. \tag{111}$$

Consider next the first term on the rhs of Eq.(111), i.e.,

$$D_n \equiv \frac{C}{2i\pi} \oint \frac{(z_3 - z)^{-1/2}}{z^{n+1}} \mathrm{d}z. \tag{112}$$

One notices first that the contour of integration in Eq.(112) may be replaced by the piecewise contour $\mathcal{H}(n)$ of the Hankel type (cf. Fig.3):

$$\mathcal{H}(n) = \mathcal{H}^-(n) + \mathcal{H}^+(n) + \mathcal{H}^0(n), \tag{113}$$

where

$$\begin{cases} \mathcal{H}^-(n) = \{z = w - \frac{i}{n},\ w \geq 1\} \\ \mathcal{H}^+(n) = \{z = w + \frac{i}{n},\ w \geq 1\} \\ \mathcal{H}^0(n) = \{z = 1 - \frac{e^{i\phi}}{n},\ \phi \in [-\frac{\pi}{2}, \frac{\pi}{2}\} \end{cases} . \tag{114}$$

Changing next the variable of integration in Eq.(111), $z = z_3(1+t/n)$, we have that D_n obeys

$$D_n = n^{-1/2} z_3^{-n} \frac{C z_3^{-1/2}}{2i\pi} \int_{\mathcal{H}} (-t)^{-1/2} \left(1 + \frac{t}{n}\right)^{-n-1} \mathrm{d}t \tag{115}$$

Further on, expanding the kernel

$$\left(1 + \frac{t}{n}\right)^{-n-1} = e^{-(n+1)\ln(1+t/n)} = e^{-t}\Big(1 + \frac{t^2 - 2t}{2n} + \dots\Big), \tag{116}$$

we observe that the integrand in Eq.(115) converges to $(-t)^{-1/2} e^{-t}$, which is just the kernel appearing in the Hankel's representation of the Gamma function. Turning to the limit $n \to +\infty$, and using an approximation

$$\left(1 + \frac{t}{n}\right)^{-n-1} = e^{-t}\left(1 + \mathcal{O}\left(\frac{1}{n}\right)\right), \tag{117}$$

in the integral in Eq.(115), we find eventually that

$$\begin{aligned} D_n &= \frac{C}{\sqrt{z_3}\Gamma(1/2)} \frac{1}{\sqrt{n} z_3^n} \left(1 + \mathcal{O}\left(\frac{1}{n}\right)\right) \\ &= \frac{C}{\sqrt{z_3 \pi}} \frac{1}{\sqrt{n} z_3^n} \left(1 + \mathcal{O}\left(\frac{1}{n}\right)\right) \end{aligned} \tag{118}$$

Next, using essentially the same kind of arguments, it is easy now to show that

$$\frac{1}{2i\pi} \oint \frac{\mathcal{O}(\sqrt{z - z_3})}{z^{n+1}} \mathrm{d}z = \mathcal{O}\left(\frac{1}{n^{3/2}}\right), \tag{119}$$

which implies that in the limit $n \to +\infty$, the leading behavior is given by

$$h_{n,0} \sim \frac{C}{\sqrt{z_3 \pi}} \frac{1}{\sqrt{n} z_3^n}, \tag{120}$$

which is exactly the result in Eqs.(58) to (60).

II. Asymptotical behavior of $h_{n,0}$ in the limit $n \to -\infty$. To study asymptotical behavior of the density profiles at large separations past stationary moving tracer, we proceed exactly in the same way as in the previous paragraph. First, we introduce the generating function of the form

$$M(z) = \sum_{n=-\infty}^{+\infty} h_{-n,0} z^n \tag{121}$$

Since $M(z)$ obeys $M(z) \equiv N(1/z)$, one immediately has that $M(z)$ is given explicitly by

$$\begin{aligned} M(z) &= \frac{(1-z)\Big(A_1 h(\mathbf{e_1}) + \rho_s(A_1 - A_{-1})\Big) + z(z-1)\Big(A_{-1} h(\mathbf{e_{-1}}) - \rho_s(A_1 - A_{-1})\Big)}{A_1 \sqrt{(z - z_1^{-1})(z - z_2^{-1})(z - z_3^{-1})(z - z_4^{-1})}} \\ &+ h(\mathbf{e_2}) \left(\sqrt{\frac{(z - z_2^{-1})(z - z_3^{-1})}{(z - z_1^{-1})(z - z_4^{-1})}} - 1 \right). \end{aligned} \tag{122}$$

Hence, $M(z)$ is an analytic function in the annular region centered around O, of inner radius z_3^{-1} and the outer radius z_2^{-1}.

Now, the leading singularity for $M(z)$ is in the vicinity of $z = z_2^{-1}$. As explained in the text, the nature of this singularity z_2^{-1} is different depending on whether the number of particles in the monolayer is explicitly conserved or not.

A. Non conserved particles number. In this case $z_2^{-1} \neq 1$ and hence, in the vicinity of $z = z_2^{-1}$, the generating function $M(z)$ behaves as

$$M(z) = E(z_2^{-1} - z)^{-1/2} - h(\mathbf{e_2}) + \mathcal{O}(\sqrt{z_2^{-1} - z}), \tag{123}$$

where E is a constant, defined by

$$\begin{aligned} E &\equiv \sqrt{z_2} \frac{(1 - z_2^{-1})\Big(A_1 h(\mathbf{e_1}) + \rho_s(A_1 - A_{-1})\Big) + z_2^{-1}(z_2^{-1} - 1)\Big(A_{-1} h(\mathbf{e_{-1}}) - \rho_s(A_1 - A_{-1})\Big)}{\sqrt{8A_2}} \\ &\times \left\{ \left(\frac{\alpha/2 - A_2}{A_1} \right)^2 - \frac{A_{-1}}{A_1} \right\}^{-1/4} \end{aligned} \tag{124}$$

Inverting Eq.(123), we find then the following asymptotical result:

$$h_{-n,0} \sim \frac{E\sqrt{z_2}}{\sqrt{\pi}} \frac{1}{\sqrt{|n|}} \frac{1}{z_2^n}, \tag{125}$$

which corresponds to our Eqs.(61) to (63).

B. Conserved particles number. In this case, $z_2^{-1} = 1$ and consequently, one has that $M(z)$ admits the following expansion in the vicinity of $z = z_2^{-1}$,

$$M(z) = -h(\mathbf{e_2}) + G(1-z)^{1/2} + \mathcal{O}\Big((1-z)^{3/2}\Big), \tag{126}$$

where G is the constant given by

$$G \equiv \frac{1}{2}\left(\frac{\Big(A_1 h(\mathbf{e_1}) + \rho_s(A_1 - A_{-1})\Big) - \Big(A_{-1} h(\mathbf{e_{-1}}) - \rho_s(A_1 - A_{-1})\Big)}{\sqrt{A_2(A_1 - A_{-1})}} + h(\mathbf{e_2})\sqrt{A_1 - A_{-1}}A_2\right) \tag{127}$$

Inverting Eq.(126), we find that in the conserved particles number case, the asymptotical behavior of the density profile at large separation past the stationary moving tracer particle obeys

$$h_{-n,0} = -\frac{G}{\pi n^3}\left(\frac{1}{2} + \frac{3}{16n} + \mathcal{O}\left(\frac{1}{n^2}\right)\right), \tag{128}$$

i.e. the behavior described by our Eqs.(64) and (65).

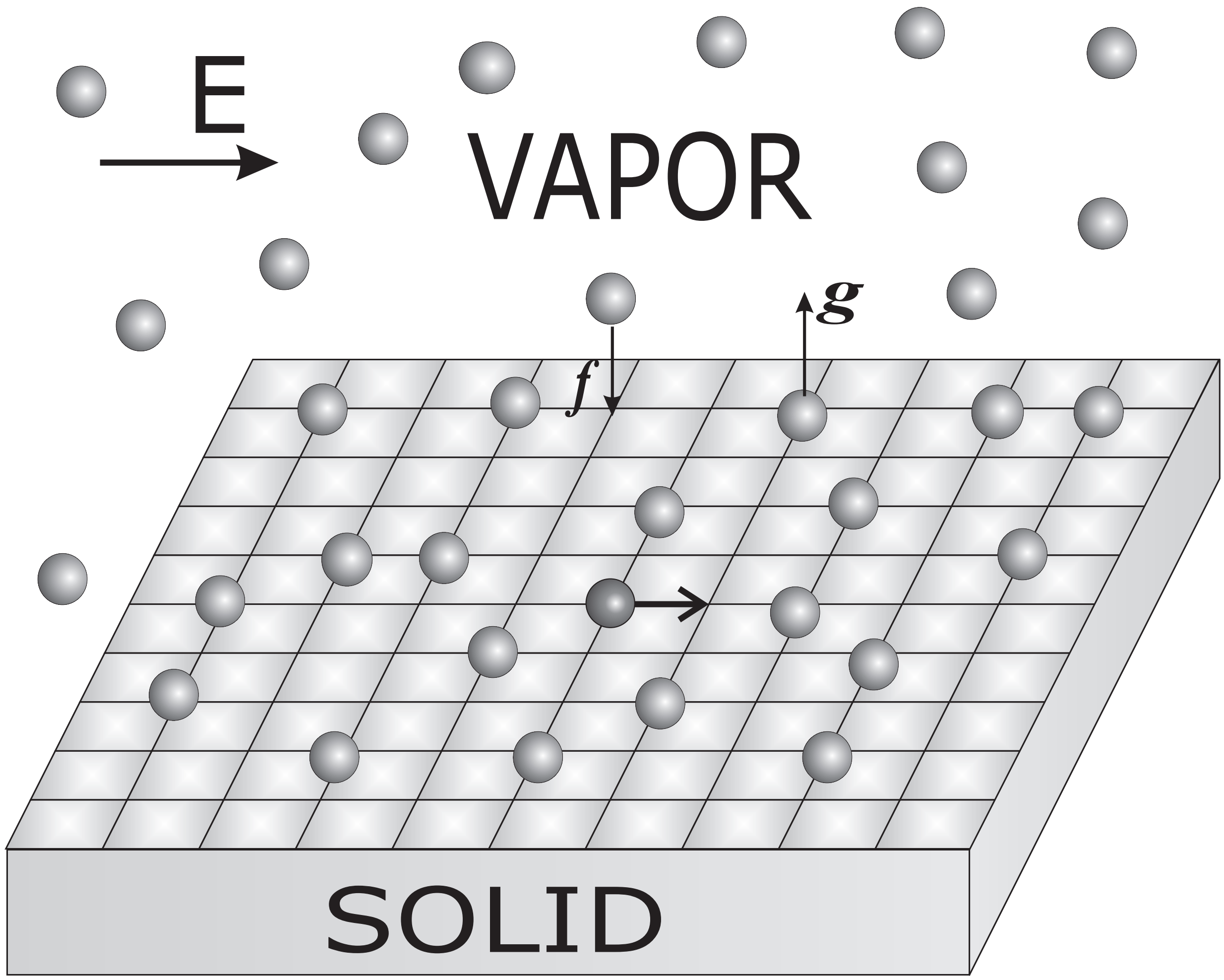

Figure 1: PRB, Bénichou et al.

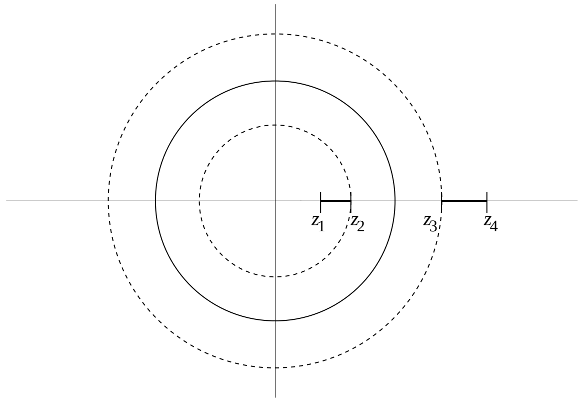

Figure 2: PRB, Bénichou et al

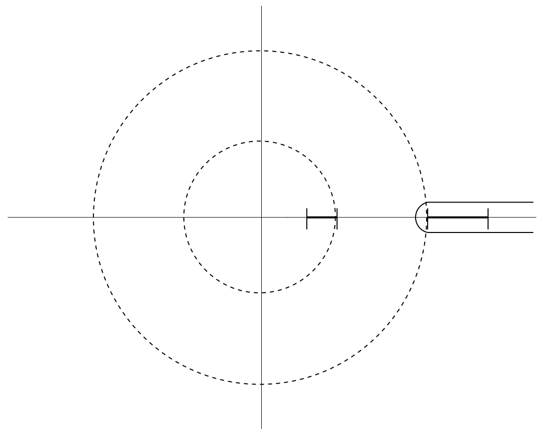

Figure 3: PRB, Bénichou et al